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The $\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3$ Ferroelectromagnetic Ceramics in a View of Possibilities to be Used for Electric Transducers

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The $\text{PbFe}_{1/2}\text{Nb}_{1/2}\text{O}_3$ (PFN) ceramics can be obtained by a lot of ways. There are a lot of methods and techniques to synthesize the PFN powder such as: reactions in the solid phase by one-stage or two-stage synthesizing method (the columbite method), molten salt synthesis, sol-gel, co-precipitation method and sintering of compacts from a loose mixture of powders. Each of them requires accuracy and precision to conduct a technological process, aiming at obtaining a product with the optimum parameters. In the work PFN specimens were obtained by one-stage synthesis methods using two different powder synthesizing techniques: by calcining (sintering of a loose mixture of the PFN powder) and as a result of sintering of pressed compacts from the PFN powder mixture at high temperature (pressing). The obtained specimens were subjected to X-ray and micro-structural examinations and tests of internal friction and dielectric properties. The tests showed that the ceramics obtained by the powder calcining method has better applied properties.

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1. Introduction

The $\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3$ ferroelectromagnetic ceramics, called shortly PFN, belong to a family of materials with a perovskite structure with the $\text{A}(\text{B}'\text{B}'')\text{O}_3$ general formula, where in positions A Pb ions substitute themselves, and in octahedral positions B' and B'' ions of iron and niobium in a random way [1–3]. The PFN material is characterized by two ordered antiferromagnetic and ferroelectric subsystems. A change from the paramagnetic to ferroelectric phase takes place at about -130°C temperature, whereas a change from the paraelectric to ferroelectric phase occurs at about 110°C temperature [4, 5].

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A technological process of the PFN ferroelectromagnetic ceramics (among others a method and a way of powder synthesizing, a type of refinement and compacting of a powder, a selection of sintering conditions, etc.) has a significant influence on a crystalline structure and a ceramics microstructure. The final properties of a material such as density, the optimum applied parameters and their stability in time depend on perfection of the crystalline structure and microstructure. In a case of the mass production an application of the PFN ceramics powder technology is the most widely used. However, during the PFN production there are a lot of barriers which impede to obtain a material with good parameters. They are: high electric conduction, or there is presence of a second non-ferroelectric pyrochlore phase besides the perovskite ferroelectric phase in the technological process.

An aim of this work was to develop a production technology of the PFN ceramics with the best applied parameters. Test results of the PFN ceramics obtained by two methods are presented. In the first a synthesis was conducted as a result of sintering of simple oxides pressed into compacts, whereas in the second synthesizing was made by the calcining of mixed oxides (CMO).

2. Experiment

The $\text{Pb}(\text{Fe}_{1-x}\text{Nb}_x)\text{O}_3$ was obtained by two methods. In the first synthesizing took place as a result of sintering of compacts from a mixture of PbO , Fe_2O_3 and Nb_2O_5 oxides in the conditions $T_{\text{synth}} = 850^\circ\text{C}/t_s = 4 \text{ h}$ (PFN1), whereas in the second CMO was used in the conditions $T_s = 850^\circ\text{C}/t_s = 4 \text{ h}$ (PFN2). Compacting of the synthesized PFN powders was made by the free sintering (FS) method. For PFN1 as a result of sintering of compacts twice in the conditions: $T_s = 950^\circ\text{C}/t_s = 4 \text{ h}$ and $T_s = 1050^\circ\text{C}/t_s = 4 \text{ h}$, whereas for PFN2 in the conditions: $T_s = 1050^\circ\text{C}/4 \text{ h}$.

The X-ray measurements were made on a polycrystalline diffractometer of the Philips firm with a Cu lamp and the graphite monochromator. Microstructure examinations of the specimen fractures were made on a SEM HITACHI S-4700 scanning microscope. Measurements of the $Q^{-1}(T)$ internal friction and $f_r(T)$ resonance frequency were made by an automatic relaxator of acoustic frequencies of a RAK-3 type, and dielectric tests were made on a capacity bridge of a QuadTech 1920 Precision LCR Meter type for $\nu = 1 \text{ kHz}$. Real density was determined by a measurement of mass and specimen volume, and $\rho_{\text{rentg}} = 8.457 \text{ g/cm}^3$ was taken as the X-ray density [6, 7].

Percentage content of the perovskite phase was calculated according to formula (1):

$$P_{\text{perow}} = 100 \left(\frac{I_{110}}{I_{110} + I_{222}} \right) [\%], \quad (1)$$

where I_{110} and I_{222} are intensities of the (110) perovskite and (222) pyrochlore diffraction lines.

3. The results and their discussion

In the PFN ceramics the pyrochlore non-ferroelectric phase is formed first (at 500°C), whereas the perovskite ferroelectric phase begins to appear above that temperature. Coexistence of those phases is obtained at the temperature of 700°C, whereas the pure perovskite phase at 750°C [6]. The CMO method used in the one-stage synthesis enables to minimize the pyrochlore phase during the PFN ceramics production [8]. The X-ray examinations for ceramics PFN1 and PFN2 are presented in Fig. 1a and Fig. 1c, respectively. Coexistence of the perovskite and pyrochlore phase can be seen clearly in a case of the PFN1 ceramics and small amounts of the non-perovskite pyrochlore phase in a case of PFN2. Percentage content of the P_{perov} perovskite phase for PFN1 and PFN2 is 94.58% and 96.77%, respectively.

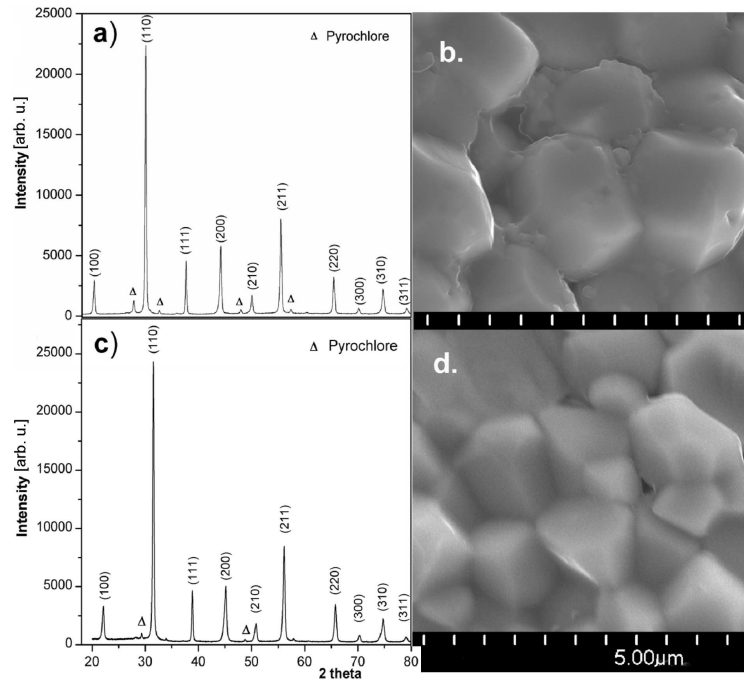


Fig. 1. X-ray spectra and fracture microstructure photographs ($\times 10000$) PFN ceramic specimens: (a, b) for specimen PFN1 and (c, d) for specimen PFN2.

In spite of presence of the polychlore phase the PFN ceramics obtained is characterized by the fine-grained structure with low porosity and densely packed grain. The microstructure examinations showed that lower average size of grains had ceramics obtained by the CMO technology (Fig. 1d).

Diagrams of relationships of electric permittivity and dielectric losses in the temperature function for the tested compositions of the PFN ceramics are pre-

TABLE

Presentation of parameters of the PFN ceramic specimens in question (heating cycle).

	$\rho_{\text{exp}} \times 10^3$ [kg/m ³]	$\rho_{\text{exp}}/\rho_{\text{theor}}$ $\times 100$ [%]	T_m [K]	ε_r	$(\tan\delta)_{Tr}$	ε_m	$(\tan\delta)_{Tm}$
PFN1	7.636	90.29	114	2900	0.052	8420	0.71
PFN2	8.180	96.72	110	2890	0.062	7530	0.14

sented in Fig. 2. The PFN ceramics obtained by the compact synthesizing method has slightly higher values of electric permittivity in comparison with specimen PFN2 obtained as a result of powder calcining. However, the conducted temperature tests of dielectric losses (Fig. 2) show that the dielectric losses for specimen PFN1 are much higher than for specimen PFN2, with simultaneous high tendency to their increase with the temperature (Table).

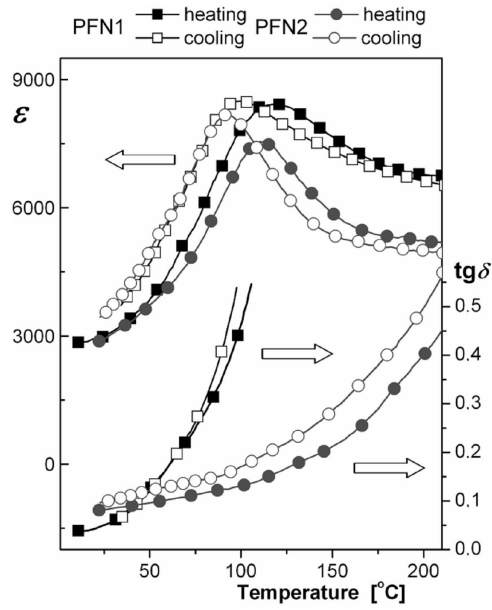


Fig. 2. Diagrams of the $\varepsilon(T)$ and $\tan\delta(T)$ relationships for the PFN ceramic specimens in question.

Figure 3 presents temperature relationships of the $E(T)$ Young modulus and $Q^{-1}(T)$ internal friction obtained for the tested specimens of ceramics PFN1 and PFN2. It is clearly visible that ceramics PFN1 has higher values of mechanical losses (the internal friction) than ceramics PFN2. It correlates with the relationships observed in a case of the $\tan\delta$ dielectric losses. Analyzing courses of the $E(T)$

relationships we can see that in the whole temperature ranges, the E Young modulus values for the specimens of ceramics PFN1 are much lower than for PFN2. At room temperature the E Young modulus values for the PFN ceramics are: $E = 20$ GPa for PFN1 and $E = 36$ GPa for PFN2, respectively.

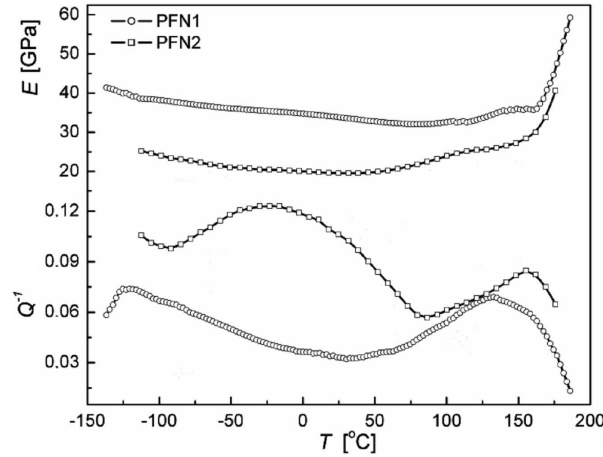


Fig. 3. Diagrams of the $Q^{-1}(T)$ and $E(T)$ relationships for the PFN ceramic specimens in question.

4. Conclusions

Synthesizing of the $Pb(Fe_{1/2}Nb_{1/2})O_3$ ceramics by a sintering method of simple oxides in the solid phase causes that an undesired pyrochlore phase may appear also besides the perovskite ferroelectric phase. Use of a PFN loose powder in the synthesizing technology reduces the above problems significantly which has an advantageous influence on the final applied parameters of the ceramics.

In a case of one-stage production of the $Pb(Fe_{1/2}Nb_{1/2})O_3$ ceramics it seems to be justifiable to conduct technology by the CMO method (calcining of a powder mixture) to obtain the appropriate crystalline structure and correct microstructure which has a direct influence on the final applied parameters of the ceramics. Improvement of the parameters of this material can be made further by modifying the CMO method introducing two-stage synthesizing of the PFN component powders (the columbite method).

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